

Role of commensurate and incommensurate low-energy excitations in the paramagnetic to hidden-order transition of URu₂Si₂

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We report low-energy inelastic neutron scattering data of the paramagnetic (PM) to hidden-order (HO) phase transition at $T_0 = 17.5$ K in URu₂Si₂. While confirming previous results for the HO and PM phases, our data reveal a pronounced wavevector dependence of quasielastic scattering across the phase transition. Temperature scans at a small energy transfer of 0.5 meV establish an abrupt step-like suppression of the excitations at $\vec{Q}_1 = (1.44, 0, 0)$ below T_0 , whereas excitations at $\vec{Q}_0 = (1, 0, 0)$, associated with large-moment antiferromagnetism (LMAF) under pressure, are enhanced with a pronounced peak at T_0 . This is the behavior expected of a “super-vector” order parameter with nearly degenerate components for the HO and LMAF leading to nearly isotropic fluctuations in the combined order-parameter space.

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For over twenty years one of the most prominent unexplained properties of f -electron materials has been the phase transition in URu₂Si₂ at $T_0 \approx 17.5$ K into a state known as ‘hidden order’ (HO) [1–4]. The discovery of the HO was soon followed by the observation of a small antiferromagnetic moment (SMAF), $m_s \approx 0.01 - 0.04 \mu_B$ per U atom [5], then believed to be an intrinsic property of the HO. The observation of a large-moment antiferromagnetic phase (LMAF) with $m_s \approx 0.4 \mu_B$ [6] under pressure consequently prompted intense theoretical efforts to connect the LMAF with the SMAF and the HO. However, following μ SR and NMR measurements [7, 8], recent Larmor and magnetic neutron diffraction allowed us to quantitatively show the parasitic nature of the SMAF [9]. Studies of the pressure–temperature phase diagram of URu₂Si₂ consistently establish the existence of a bicritical point, which implies that HO and LMAF break *different* symmetries [9–13]. These properties point to exotic scenarios of the HO, such as incommensurate orbital currents [14], helicity order [15], multipolar order [16, 17], or a spin-nematic state [18, 19].

Inelastic neutron scattering has been essential for gaining microscopic insight into the nature of the HO (see e.g. [20, 21]). The existence of commensurate and incommensurate excitations in the HO at $\vec{Q}_0 = (1, 0, 0)$ and $\vec{Q}_1 = (1.44, 0, 0)$, respectively, had been known for a while [5]. However, their precise relationship to the HO is an open issue of central importance, and consequently there has been controversy on which excitations

“drive” the transition. Namely, the incommensurate excitations at \vec{Q}_1 are reported to be gapped in both the HO and LMAF phase, with gaps of Δ approximately 4 meV and 8 meV, respectively [22, 23], but gapless in the PM phase [24]. The closing of the gap has been quantitatively linked to the specific heat jump at T_0 [24], suggesting that the incommensurate fluctuations drive the PM-HO transition. In contrast, it has recently been suggested that the commensurate excitations at \vec{Q}_0 drive the PM-HO transition, based on the observation that they occur only in the HO phase (Δ approximately 2 meV) while being absent in the PM and LMAF phase [21, 23].

In this Letter we present compelling evidence that the *commensurate* fluctuations at \vec{Q}_0 , which can be interpreted as longitudinal LMAF fluctuations, are most closely connected to the PM-HO transition. Further, the LMAF and HO fluctuations are likely interrelated. Detailed temperature scans across the HO-PM phase transition at low energies show qualitatively different behavior at \vec{Q}_0 and \vec{Q}_1 . At the incommensurate \vec{Q}_1 position the gap is filled abruptly upon heating at T_0 , i.e., the low-energy excitations are suppressed in a step-like fashion upon entering the HO phase. In contrast, scans at the commensurate \vec{Q}_0 position show for the first time that the low-energy excitations are enhanced across a considerable temperature range. Most importantly, they peak at T_0 , suggesting that these fluctuations become *almost critical* at the PM-HO transition in addition to the expected critical fluctuations of the hitherto unidentified HO parameter. This is not expected in a standard sce-

nario of competing order parameters for HO and LMAF, which break different symmetries. However, this is consistent with nearly isotropic fluctuations of a super-vector order parameter which consists of components for both HO and LMAF. Isotropy in this order-parameter space would imply an emergent symmetry between both orders, which may be tested experimentally.

Experimental.— The single crystal studied was grown by means of an optical floating-zone technique at the Amsterdam/Leiden Center. High sample quality was confirmed via X-ray diffraction and detailed electron probe microanalysis. The mosaic spread is less than 1° . Samples prepared from this ingot showed good resistance ratios (20 for the c axis and ≈ 10 for the a axis) and a high superconducting $T_c \approx 1.5$ K. The magnetization of the large single crystal agreed very well with data shown in Ref. [25] and confirmed the absence of ferromagnetic inclusions. Most importantly, in our neutron scattering measurements we found an antiferromagnetic moment $m_s \approx 0.012 \mu_B$ per U atom [9], which matches the smallest moment reported so far [26].

Inelastic neutron scattering measurements were carried out at the cold triple-axis spectrometer Panda at FRMII. The sample was mounted on a Cd-shielded Cu holder and oriented with $(h0l)$ as the horizontal scattering plane. Panda was used in W-configuration with vertically and horizontally focusing monochromator and analyser and no collimation. The final wavevector was kept fixed at 1.55 \AA^{-1} . Higher-order harmonics were removed from the scattered beam by a liquid-nitrogen cooled Be filter and monitor correction for higher order neutrons was included. The temperature evolution of the low-energy excitations of URu_2Si_2 at commensurate \vec{Q}_0 and incommensurate \vec{Q}_1 was studied by low-energy scans at different temperatures. Most importantly, detailed temperature scans at $E = 0.5 \text{ meV}$ were carried out at each position.

Energy scans.— Fig. 1 shows typical energy scans for \vec{Q}_0 and \vec{Q}_1 . At both positions quasielastic scattering is found for temperatures above T_0 . At T_0 gaps are opening up and with further decreasing T the intensities of the excitations increase while the gaps widen. The spectrum is clearly gapped at low temperatures. At 3 K, low-energy excitations are detected at 2 meV at \vec{Q}_0 and at 4.5 meV at \vec{Q}_1 . Fig. 2 shows that the quasielastic scattering develops in different ways at \vec{Q}_0 and \vec{Q}_1 as T_0 is approached from above. The intensity of the commensurate quasielastic scattering significantly increases at energies below about 2 meV. In stark contrast, the incommensurate quasielastic scattering stays almost constant.

The quasielastic scattering in Fig. 2 consists of the contribution from the dynamic structure factor and a background. The data may be well fitted with a model for the dynamic structure factor in which the imaginary part of

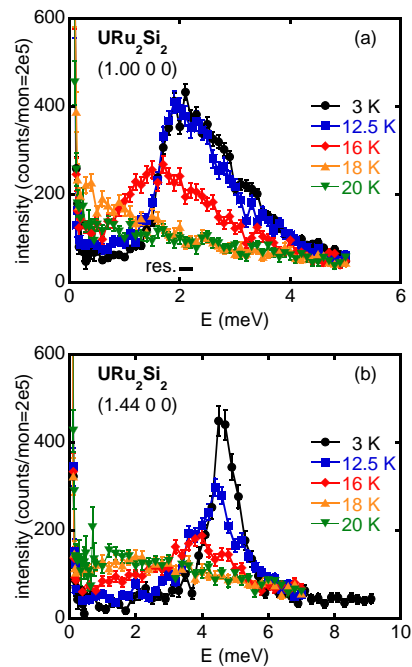


FIG. 1: Low-energy excitations. At low temperature T in the hidden order phase excitations are gapped. Excitations are seen (a) above $\Delta \approx 2 \text{ meV}$ at the commensurate $(1, 0, 0)$ position and (b) above $\Delta \approx 4 \text{ meV}$ at the incommensurate $(1.44, 0, 0)$ position. Both gaps close at the transition to paramagnetism ($T_0 = 17.5 \text{ K}$). (mon=2e5 $\hat{=}$ $\sim 2 \text{ min}$ counting time. The 'res.' bar indicates the resolution.)

the dynamic susceptibility is expressed by a Lorentzian

$$S(\vec{q}, \omega) = \left(\frac{1}{1 - e^{-\hbar\omega/k_B T}} \right) \frac{\hbar\omega\chi'_{\vec{q}}\Gamma_{\vec{q}}}{\hbar\omega^2 + \Gamma_{\vec{q}}^2}$$

Here, $\chi'_{\vec{q}}$ is the real part of the static susceptibility, and $\Gamma_{\vec{q}}$ denotes the quasielastic linewidth. The background as determined in energy scans at 20 and 3 K at $(1.2, 0, 0)$ was found to be constant in the parameter range and essentially independent of T . The background is thereby believed to include the magnetic continuum recently reported in Ref. 21.

At the commensurate position $\Gamma_{\vec{Q}_0}$ reduces significantly from 1.5 meV at 20 K to 1.1 meV at 18 K. The static susceptibility $\chi'_{\vec{Q}_0}$ does not become critical when approaching T_0 , but is enhanced by a factor of 1.5 upon cooling from 20 to 18 K. This contrasts the critical enhancement of $\chi'_{\vec{Q}_0}$ reported by Mason *et al.* [27], which, however, was based on an analysis in terms of a spin-wave fit for data above T_0 . Perhaps most importantly, our data agrees with the results reported by Bourdarot *et al.* [21], who observed a reduction of $\Gamma_{\vec{Q}_0}$ (dropping from 1.4 meV at 20 K to 1.2 meV at 18 K) and an enhancement of $\chi'_{\vec{Q}_0}$ by a factor of 1.6.

The properties at the incommensurate position, \vec{Q}_1 , contrast those recorded at \vec{Q}_1 , where the quasielastic

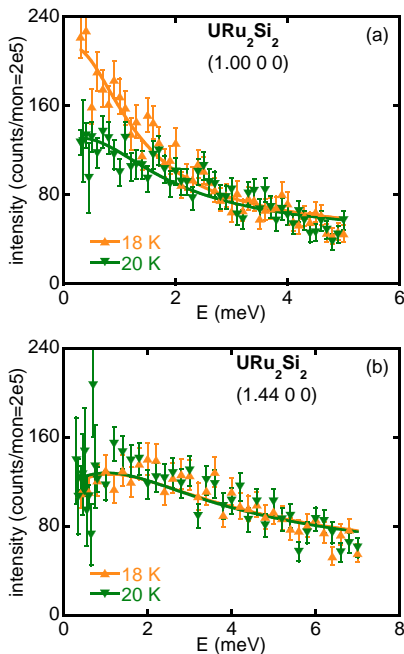


FIG. 2: Quasielastic scattering above T_0 . Data above 0.3 meV are fitted with a standard model for the dynamic structure factor containing a Lorentzian and a constant background (see text). (a) At the commensurate $(1, 0, 0)$ position quasielastic scattering is significantly enhanced towards T_0 and $\Gamma_{\vec{Q}_0}$ is reduced from 1.5 meV at 20 K to 1.1 meV at 18 K. (b) At the incommensurate $(1.44, 0, 0)$ position, however, there is almost no difference in the quasielastic scattering at 20 and 18 K, and the linewidth is quite large ($\Gamma_{\vec{Q}_1} = 2.6$ meV).

scattering at 20 and 18 K is essentially the same and the linewidth quite large ($\Gamma_{\vec{Q}_1} = 2.6$ meV). This qualitatively agrees with data by Broholm *et al.* [20] where the values for $\Gamma_{\vec{Q}_1}$ were reported to be between 5 and 6 meV.

Temperature scans.— While the spin fluctuations are truly critical neither at \vec{Q}_0 nor at \vec{Q}_1 , we have observed significant differences between the low-energy excitation spectra at \vec{Q}_0 and \vec{Q}_1 when approaching the onset of hidden order at T_0 . For both wave vectors these differences are best seen in temperature scans at $E = 0.5$ meV (Figure 3). At the incommensurate position, \vec{Q}_1 , the gap opens in an almost step-like fashion at T_0 . This agrees with data by Wiebe *et al.* [24], where no enhancement of the low- E excitations was observed when approaching T_0 from above. However, our data shows a much sharper decrease of scattering intensity just below T_0 : Wiebe *et al.* had studied an energy transfer $E = 0.25$ meV where a full suppression of the excitations was seen below 13 K, whereas our scan at $E = 0.5$ meV reaches the background level already at 16 K. In strong contrast, at the commensurate position, \vec{Q}_0 , the low-energy excitations are only fully suppressed below 11 K. Most importantly, the low-energy excitations are enhanced from about 12 to 24 K

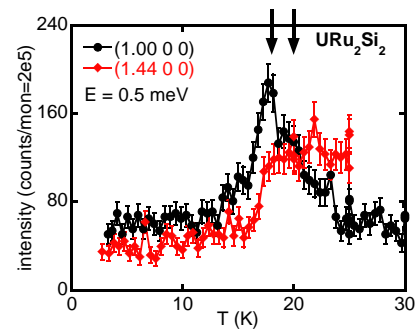


FIG. 3: Temperature dependence of the low-energy excitations around the hidden-order to paramagnetic phase transition. At the commensurate $(1, 0, 0)$ position the gap is filled across a considerable temperature range around T_0 . Low-energy excitations at $E = 0.5$ meV are strongly enhanced and peak precisely at T_0 . At the incommensurate $(1.44, 0, 0)$ wavevector the gap is filled much more abruptly at T_0 and the $E = 0.5$ meV excitations do not show any additional enhancement. Arrows denote the positions of energy scans above T_0 shown in Fig. 2.

and peak precisely at T_0 .

Discussion.— The detailed temperature scans suggest an intimate relation between the PM-HO phase transition and the commensurate excitations at \vec{Q}_0 . The latter show a softening towards T_0 , much like the critical fluctuations of a second-order phase transition. In marked difference, the incommensurate excitations at \vec{Q}_1 do not show any significant softening but persist at essentially constant intensity when T_0 is approached from higher temperatures. The opening of the gap in these incommensurate excitations may in turn be interpreted as a simple *consequence* of the onset of the HO. This view is not incompatible with the proposal [24] that the incommensurate excitations are mainly responsible for the magnitude of the specific-heat anomaly.

The commensurate fluctuations at \vec{Q}_0 – although being peaked exactly at T_0 – do not become critical, i.e., the corresponding static susceptibility does not diverge. The latter is only expected if the magnetic order becomes static at \vec{Q}_0 below T_0 , which would be the case in the pressure-induced LMAF phase, but not in the HO phase. What is then the role of the \vec{Q}_0 magnetic fluctuations?

It is instructive to discuss the interplay of hidden order and magnetism using the order-parameter language. If HO and magnetism would simply represent competing order parameters ψ_{HO} and ψ_{AF} , with ordering wavevectors \vec{Q}_{HO} and \vec{Q}_{AF} , respectively, an enhancement of the HO would lead to a suppression of magnetism and vice versa. In particular, it would be expected that magnetic fluctuations are suppressed instead of enhanced when approaching the HO transition. Moreover, one would not expect that the HO couples to the magnetism in a wavevector-selective manner: To lowest order the allowed coupling in a Landau functional is of the form

$|\psi_{\text{HO}}|^2|\psi_{\text{AF}}|^2$, which does not require any relationship between \vec{Q}_{HO} and \vec{Q}_{AF} . Therefore, a standard scenario of competing orders with differing symmetries, inferred from the parasitic nature of the small-moment antiferromagnetism and the temperature-pressure phase diagram [9], does not easily account for our data.

This prompts us to invoke a closer relationship between HO and magnetism. A specific proposal along these lines was recently made by Haule and Kotliar [17, 28]. The central idea, supported by microscopic calculations [17], is that ψ_{HO} and ψ_{AF} may be treated as components of a *common* super-vector order parameter. This implies that the system is in the vicinity of a point with higher symmetry, where HO and magnetism are degenerate. Approaching the ordering transition at T_0 in turn will lead to a concomitant enhancement of *both* HO and magnetic fluctuations, corresponding to nearly isotropic fluctuations in order-parameter space, until – very close to the PM-HO transition – the magnetic fluctuations are cut-off, consistent with our data. In the simplest case, this scenario suggests $\vec{Q}_{\text{HO}} = \vec{Q}_{\text{AF}} = (0, 0, 1)$. Therefore it will be crucial to search for the proposed [17] hexadecapolar order at wavevector $(0, 0, 1)$ in the HO phase, and also to detect its fluctuations at ambient pressure.

Conclusions.— We have shown a strong link between commensurate magnetic fluctuations at \vec{Q}_0 and the PM-HO transition in URu_2Si_2 . Temperature scans of the low-energy excitations at the commensurate \vec{Q}_0 and the incommensurate \vec{Q}_1 positions show qualitatively different behaviour across the transition, with the former being strongly enhanced towards the PM-HO transition temperature T_0 . Our observations put strong constraints on theoretical models for the HO state; they point at a common, nearly isotropic, order-parameter space involving both HO and LMAF order parameters [28].

As a consequence we predict for high-pressure neutron scattering experiments of the low-energy excitations at \vec{Q}_0 and \vec{Q}_1 near the PM-LMAF transition, that the fluctuations at \vec{Q}_0 become stronger for increasing pressure at $T_0(p)$, with a clear trend to a truly critical divergence at T_N beyond the bicritical point [9–13]. At the same time, the intensity at \vec{Q}_1 is predicted to remain non-critical, step-like, at all p and T .

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